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(54) Title: FILMS OF LINEAR LOW DENSITY POLYETHYLENE HAVING IMPROVED IMPACT STRENGTH

(57) Abstract

Compression-rolled films of linear low density polyethylene have high impact strength and high tear strength in both the machine and transverse directions.

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FILMS OF LINEAR LOW DENSITY POLYETHYLENE HAVING IMPROVED IMPACT STRENGTH

This invention relates to films of linear low density polyethylene (LLDPE).

LLDPE has a molecular structure which is characterized by the substantial absence of long chain branching. In contrast, conventional low density polyethylene (LDPE) has substantial long chain branching. LLDPE also has a significantly higher melting point (typically 120-135°C) than LDPE (typically 105-115°). LDPE is sometimes referred to as high pressure polyethylene because it is produced at high pressures. LLDPE, on the other hand, is produced commercially at low pressures in a gas phase process. However, LLDPE may also be produced in a liquid phase solution process.

Various alpha-olefins are typically copolymerized with ethylene in producing LLDPE. The alpha-olefins , which have 3 to 15, preferably 4 to 8, carbon atoms, are present in the polymer in an amount of up to about ten weight percent. The comonomer influences the density of the polymer, which is less than 0.94 grams per cubic centimeter, and preferably is from about 0.915 to 0.935 g/cc.

This invention provides films of LLDPE having substantially improved impact strength compared to conventional films of LLDPE. The films of the invention also have good tear strength in both the transverse and machine directions.

The films of this invention are prepared by compression rolling films of LLDPE. The term "compression rolling," as used herein and as

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understood in the art, refers to the process of passing a plastic film through the nip of a pair of rolls at a temperature below the softening point of the film at a pressure which is sufficient to reduce the thickness of the film at least five percent, preferably at least fifty percent. The length of the film is increased by an amount corresponding to the reduction in thickness. For example, a film whose thickness has been reduced fifty percent by compression rolling has a length which is about twice as great as its original length. Compression rolling is described in U.S. Patents 3,504,075 and 4,085,187, which are incorporated herein by reference.

These patents teach that virtually any polymer may be compression rolled, but compression rolling may adversely affect certain properties of a film to make it unacceptable. For example, as taught in an article by Williams and Morrison in the December 1971 issue of SPE Journal (Vol. 27), pp. 42-48, compression rolling of high density polyethylene (HDPE) and polypropylene (PP) films decrease their tear stength in the machine direction. As shown in Table 2 of the article, rolled films of HDPE and PP having a thickness of one mil had tear strengths in the machine direction of only 24 grams and 5 grams, respectively. Hence, compression rolling of films of HDPE and PP makes them "splitty" and reduces their impact strength, which severely limits their commercial utility.

An opposite phenomenon is observed with LDPE. As shown in Table 2 of the Williams article, rolling improves the tear strength of films of LDPE in the machine direction, but it reduces their tear

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strength in the transverse direction. In fact, the tear stength of rolled LDPE film in the transverse direction is so low that LDPE film is difficult to roll at high speeds because it tends to tear in the transverse direction as it is drawn from the rolling mill.

This invention is based on the discovery that compression rolling of films of LLDPE substantially improves the impact strength of the films and ensures that they have good tear strength in both the machine and transverse directions. This result is surprising and unexpected in view of the effect of compression rolling on the impact strength and tear strength of films of other polyolefins. For example, the rolled films of LLDPE of this invention have notched Elmendorf tear strengths (ASTM D-1922) of at least 300 (in preferred embodiments at least 350) grams per mil in both the machine and transverse directions, which cannot be said for other rolled polyolefin films. The films of this invention also have a falling dart impact strength (ASTM D-1709) of at least 500 grams per mil when the dart is dropped from a height of 8.75 inches, and at least 160, preferably at least 180, grams per mil when the dart is dropped from a height of 26 inches. The impact strength of conventional LLDPE film is much lower.

The compression rolling is preferably carried out with semiboundary lubrication, as taught in U.S.P. 4,085,187, with the preferred lubricant being water. The film is preferably rolled at a temperature of about 15 to 80 ^{o}C, more preferably 20 to 55^{o}C. The thickness of the film is preferably reduced between about 50 and 90

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percent. The final thickness of the films of this invention is preferably between about 0.5 and 6 mils, more preferably between one and three mils. In addition to high impact and tear strength, the films of this invention have other excellent properties compared to conventional films, including stiffness, tensile strength, and water vapor transmission rate.

Other polymers can be blended or coextruded with the LLDPE to provide compression-rolled films with tailored properties. Such other polymers include other polyolefins, such as HDPE (i.e. polyethylene having a density greater than 0.94 g/cc.), PP and copolymers of ethylene or propylene with minor amounts of other ethylenically unsaturated monomers. The proportion of LLDPE blended or coextruded with other polymers is preferably at least 25, more preferably at least 50, percent by weight.

In the following examples, which illustrate the invention, "MD" means machine direction; "TD" means transverse direction, "CNT" means cannot tear; "MI" means melt index; "NT" means not tested; "tear strength" means notched Elmendorf tear strength (ASTM D-1922) measured in grams per mil; and "impact strength" is measured by the dart drop test of ASTM D-1709 in grams per mil, with the dart being dropped from a height of 8.75 or 26 inches.

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EXAMPLES 1 -4
AND COMPARATIVE EXAMPLES A-J

In Examples 1-4 and F, H and J, cast films consisting of a single polymer, identified in the following Table, were compression rolled at ambient temperature. The ratio of the thickness of the film before rolling to the thickness after rolling is shown in parentheses in the Table. The properties of the compression-rolled films are compared to representative cast and blown unrolled films (Examples A, B, C, D, E, G and I) in the Table.

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TABLE

Polymer	Film Type	Thickness Mils	Tear Strength MD	Tear Strength TD	Impact Strength 8.75"	Impact Strength 26"	Example
LLDPE, MI=6, density = 0.919	Cast	0.9	370	740	270	90	A
	Rolled (4.9)	1.3	480	760	500	185	1
	Blown	1.0	130	380	NT	130	B
	Rolled (4.1)	1.5	490	CNT	NT	160	2
LLDPE, MI=2.5, density = 0.935	Cast	1.1	50	870	180	50	C
	Rolled (5.0)	1.3	385	485	500	225	3
	Blown	1.0	80	340	NT	65	D
	Rolled (4.3)	1.4	500	630	NT	230	4
HDPE, MI=6, density = 0.960	Cast	1.1	16	50	130	NT	E
	Rolled (5.7)	1.1	40	270	85	NT	F
PP, MI=6.5, density = 0.897	Cast	1.1	33	67	80	NT	G
	Rolled (3.0)	1.5	10	CNT	60	NT	H
LDPE, MI=2.0 density = 0.928	Cast	1.0	150	440	80	70	I
	Rolled (4.7)	1.3	460	210	210	70	J

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These examples show that when films of LLDPE are compression rolled, the impact strength of the films is substantially improved, and the tear strengths of the films are over 350 grams per mil in both the machine and transverse directions. In comparison, when films of HDPE and PP are compression rolled, the impact strength of the films is reduced, and their tear strength in the machine direction is very low. Hence, they tend to split, which makes them unsuitable for many applications. When films of LDPE are compression rolled, the tear strength of the films in the transverse direction is reduced to less than about 250 grams per mil. Hence, they tend to tear when they are drawn from the rolling mill.

The rolled films are compared in the Table with unrolled films of comparable thickness, rather than with the films before rolling, because the thickness of the films before rolling is so much greater that a comparison with them would not be as meaningful as a comparison with films of comparable thickness. Hence, while a comparison of Examples C and 3 might appear to suggest that rolling might reduce the tear strength of LLDPE in the transverse direction, that is not believed to be the case. The very high tear strength in the transvere direction of the film of Example C is believed to be due to orientation of the film that inherently or inadvertently occurred when the film was extruded. However, such orientation also apparently caused a sharp reduction in the tear strength of the film in the machine direction, which does not occur with compression rolling of LLDPE film.

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CLAIMS

1. A compression-rolled film of linear low density polyethylene having substantially improved falling dart impact strength, substantially improved tear strength in the machine direction, and a tear strength in the transverse direction of at least 300 grams per mil.

2. The film of claim 1 having an impact strength of at least 180 grams per mil when the dart is dropped from a height of 26 inches.

3. The film of claim 1 having notched Elmendorf tear strength of at least 300 grams per mil in both the machine and transverse directions.

4. The film of claim 2 having notched Elmendorf tear strength of at least 350 grams per mil in both the machine and transverse directions.

5. The film of claim 3 having a thickness between about 0.5 and 6 mils.

6. The film of claim 3 having a thickness between about one and three mils.

7. The film of claim 5 having another polymer coextruded with the polyethylene.

8. The film of claim 5 having another polymer blended with the polyethylene, with the proportion of the linear low density polyethylene being at least fifty percent by weight of the blend.

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9. The film of claim 3 wherein the density of the polyethylene is less than about 0.94 g/cc.

10. The film of claim 9 wherein the polyethylene has copolymerized therewith up to ten weight percent of another alpha-olefin.

11. The film of claim 10 wherein the alpha-olefin has from four to eight carbon atoms.

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INTERNATIONAL SEARCH REPORT

International Application No PCT/US 84/00003

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [3]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC[3]: C 08 J 5/18; C 08 L 23/04; B 29 D 7/24

II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
IPC[3]	C 08 J; C 08 L; C 08 F; B 29 D

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [5]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [14]

Category *	Citation of Document, [16] with indication, where appropriate, of the relevant passages [17]	Relevant to Claim No. [18]
X,Y	FR, A, 2269410 (UNION CARBIDE CORP.) 28 November 1975 see claims 1,7	1
Y	EP, A, 0048227 (UNIFOS. KEMI. AB.) 24 March 1982 see claims; page 8, example 2	1
Y	US, A, 4085187 (R.H. JENKS et al.) 18 April 1978 see claims; abstract (cited in the application)	1
A	A, 400564 (CHEMISCHE WERKE HÜLS) 15 April 1966 see claims	
A	US, A, 4363904 (W.A. FRASER et al.) 14 December 1982	
A	EP, A, 0010428 (MITSUI PETROCHEMICAL) 30 April 1980	

* Special categories of cited documents: [15]

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"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search [1]	Date of Mailing of this International Search Report [2]
2nd April 1984	03 MAI 1984
International Searching Authority [1]	**Signature of Authorized Officer [20]**
EUROPEAN PATENT OFFICE	G.L.M. Kruydenberg

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ANNEX TO THE INTERNATIONAL SEARCH REPORT ON

INTERNATIONAL APPLICATION NO. PCT/US 84/00003 (SA 6490)

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 26/04/84

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Patent document cited in search report	Publication date	Patent family member(s)		Publication date
FR-A- 2269410	28/11/75	DE-A-	2519075	13/11/75
		JP-A-	50146673	25/11/75
EP-A- 0048227	24/03/82	SE-B-	422070	15/02/82
US-A- 4085187	18/04/78	None		
CH-A- 400564		None		
US-A- 4363904	14/12/82	EP-A-	0021605	07/01/81
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